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A. Liñán

Instituto Nacional de Técnica Aeroespacial
Madrid, Spain

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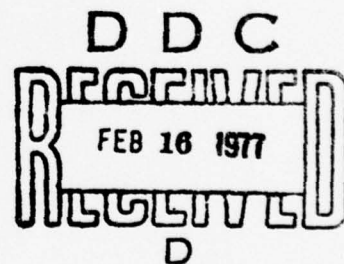
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THEORY OF HETEROGENEOUS IGNITION UNDER CONVECTIVE HEATING

by Amable Liñán

ABSTRACT

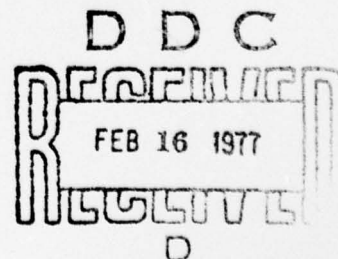
An analysis of the heterogeneous ignition of a solid, under convective heating by a hot gas, for large values of the activation energy reveals the existence of two ignition regimes. In a first regime, for large values of the frequency factor, a short reactive stage ending in thermal runaway follows a long inert stage. For small values of the frequency factor the chemical heat release can be neglected in a first stage but not in a much longer second stage when the surface temperature is close to the hot gas temperature; a critical value is found for the frequency factor below which no thermal runaway or well defined ignition time exists.

Closed form expressions for the ignition time are given for both regimes.

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I INTRODUCTION

This paper deals with the analytical description of the process of heterogeneous ignition of a solid under convective heating by a hot gas. The heat exchange between the gas and the solid is modelled by a Newtonian law, so that the heat flux from the gas to the solid is written as proportional to the difference in temperature of the gas and that of the surface of the solid. The exothermic heterogeneous reaction is considered to follow Arrhenius kinetics with an activation energy large compared with the thermal energy of the solid. The effects of interphase mass transfer are neglected here.

For a review of previous numerical and analytical work related to this model, see the review by Merzhanov and Averson¹. An asymptotic analysis for large activation energies of heterogeneous ignition under conductive heating by a hot gas was presented by Liñán and Crespo² and by Kindelán and Liñán³, who took into account the effects of gasification accompanying the heterogeneous reaction. An asymptotic analysis of ignition under convective heating by a hot gas, when an exothermic reaction

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occurs in the condensed phase has been recently completed by Niiooka and Williams⁴. The analysis presented here for heterogeneous ignition is parallel to that of Niiooka and Williams.

The chemical heat release, which is a very sensitive function of the surface temperature, becomes comparable to the convective heating only when the surface temperature approaches a value T_c . In Section III we consider the cases when the frequency factor is large enough so that T_c is intermediate between the initial solid temperature and the gas temperature. In this case there is a first long stage of inert heating followed by a short transition stage, when the chemical and convective heating are comparable, ending in a precipitous rise in temperature or thermal runaway at a well defined ignition time. In Section IV an analysis of the ignition or heterogeneous reaction process is presented for smaller values of the frequency factor when T_c becomes close to the hot gas temperature. A critical value is found for the frequency factor below which no thermal runaway occurs.

II FORMULATION

When a semiinfinite body is heated at its surface by a flux proportional to the difference in its surface temperature and that of the source, the equations describing the temperature distribution can be written in non-dimensional form as the heat conduction equation,

$$\frac{\partial \theta}{\partial t} - \frac{\partial^2 \theta}{\partial x^2} = 0 \quad x > 0, \quad t > 0 \quad (1)$$

to be solved with the initial condition

$$\theta(x, 0) = \theta_i \quad (2)$$

which is also the temperature of the solidgas from the surface at all times. In addition we have the surface condition

$$x=0, \quad t>0 : \quad - \frac{\partial \theta}{\partial x} = 1 - \theta + B e^{-\theta a / \theta} \quad (3)$$

where we have assumed that an exothermic heterogeneous reaction with Arrhenius kinetics takes place at the surface of the solid.

The temperature has been made non-dimensional with the temperature of the heat source; θ_a is the non-dimensional activation temperature. The characteristic length and time used in the nondimensional length x , time t and frequency factor B have been obtained from the thermal diffusivity, heat conduction coefficient, and overall heat transfer coefficient.

As is well known there is a relation

$$\theta - \theta_i = \frac{1}{\sqrt{\pi}} \int_0^t \frac{1 - \theta + B e^{-\theta_a/\theta}}{\sqrt{t - t_1}} dt_1 \quad (4)$$

between the surface temperature and heat transfer flux at the surface.

We shall give in this paper a description of the solution of Eq(4) for large values of θ_a , first, for fast reactions, corresponding to large values of $B \exp(-\theta_a)$, in Section III, and, second, for slow reactions, corresponding to values of $\theta_a B \exp(-\theta_a)$ or order unity or small in Section IV.

III IGNITION WITH FAST REACTIONS

For $B=0$, or for values of B and θ such that

Be^{-θ_a/θ} << (1-θ), the surface temperature history is given by θ=θ_f(t), the inert temperature value given by the solution of

$$\theta_f(t) - \theta_i = \frac{1}{\sqrt{\pi}} \int_0^t \frac{1 - \theta_f(t_1)}{\sqrt{t - t_1}} dt_1 \quad (5)$$

takes the value

$$(1 - \theta_f) / (1 - \theta_i) = e^{t \operatorname{erfc} \sqrt{t}} \quad (6)$$

For large values of the nondimensional activation temperature θ_a and values of B such that

$$1 - \theta_c = B \exp(-\theta_a / \theta_c), \quad \theta_i < \theta_c < 1 \quad (7)$$

There is an initial inert stage, as outlined by Liñán and Williams⁵, where the temperature θ takes the inert value of Eq(6), except for exponentially small terms (~exp(θ_a/θ_c - θ_a/θ)); the inert stage is followed by a short transition stage around t=t_c, where t_c is given by

$$\theta_f(t_c) = \theta_c \quad (8)$$

The transition stage ends with a thermal runaway at a well defined ignition time t_i , given by the relation

$$B = e^{-0.431} \left(\frac{\theta_{fi}^2}{\theta_a \theta'_{fi}} \right)^{1/2} e^{\theta_a / \theta_{fi}} \quad (9)$$

where

$$\theta_{fi} = \theta_f(t_i) \quad \text{and} \quad \theta'_{fi} = \frac{d\theta_f}{dt}(t_i)$$

as it can be shown through a procedure paralleling that used by Liñán and Crespo².

Notice that the characteristic time of the transition stage is of order $1/\theta_a \theta'_f(t_c)$, and becomes very large when $\theta_c \rightarrow 1$ because $\theta'_f(t_c) \rightarrow 0$; in this case the analysis leading to Eq(9) for the ignition time fails. We shall here present an analysis of the solution of the system of Eqs(1)-(3) for values of B such that the relation (7) is satisfied only for values of $\theta_c \geq 1$. In fact there is a distinguished limiting case corresponding to values of θ_c very close to 1, which we shall analyze in detail below.

IV IGNITION WITH SLOW REACTIONS

We shall describe here the solution of Eq(4) for

large values of θ_a and values of B such that

$$\delta = \theta_a B \exp(-\theta_a) \quad (10)$$

is of order unity. In this case the temperature distribution will be given by Eq(6) until t is large enough so that

$$1 - \theta_f \doteq (1 - \theta_i) / \sqrt{\pi t}$$

Small temperature increments, or order $1/\theta_a$, around $\theta=1$ suffice to multiply the reaction term by a factor e making it comparable or more important than the Newtonian heating term. We shall write Eq(4) in terms of the variable.

$$\theta = \psi / \theta_a + \theta_f \quad (11)$$

and notice that in first approximation we can write the Arrhenius exponent in the form

$$e^{-\theta_a / \theta} \doteq e^{-\theta_a e^{\psi - \theta_a(1 - \theta_i) / \sqrt{\pi t}}} \quad (12)$$

which is the correct asymptotic form of the exponent

when the reaction term can not be neglected. With this approximation Eq(4) becomes

$$\psi = \frac{1}{\sqrt{\pi}} \int_0^t \frac{-\psi + \delta e^{-\alpha/\sqrt{t_1} + \psi}}{\sqrt{t-t_1}} dt_1 \quad (13)$$

where

$$\alpha = \theta_a(1-\theta_i)/\sqrt{\pi} \quad (14)$$

The integral equation(13), which is of the Abel type, will be solved below for large values of α in the distinguished limiting case when δ is of order 1. To obtain the asymptotic solution we begin by writing it in terms of the variable

$$\sigma = t/\alpha^2 \quad (15)$$

to obtain

$$\psi(\sigma)/\alpha = \frac{1}{\sqrt{\pi}} \int_0^\sigma \frac{-\psi(\sigma_1) + \delta e^{\psi(\sigma_1) - 1/\sqrt{\sigma_1}}}{\sqrt{\sigma - \sigma_1}} d\sigma_1 \quad (16)$$

which can be inverted to yield

$$-\psi + \delta e^{\psi - 1/\sqrt{\sigma}} = \frac{1}{\alpha\sqrt{\pi}} \int_0^\sigma \frac{d\psi(\sigma_1)}{\sqrt{\sigma - \sigma_1}} \quad (17)$$

We then introduce the expansion

$$\psi = \psi_0 + \frac{1}{\alpha} \psi_1 + \dots \quad (18)$$

in Eq(17) to obtain

$$\psi_0 e^{-\psi_0} = \delta e^{-1/\sqrt{\sigma}} \quad (19)$$

and

$$\psi_1 (\psi_0 - 1) = \frac{1}{\sqrt{\pi}} \int_0^{\sigma} \frac{d\psi_0(\sigma)}{\sqrt{\sigma - \sigma_1}} \quad (20)$$

Eq(19)*, giving the first approximation for $\psi(\sigma)$, has two branches; the one with physical meaning is the lower one and it shows ψ growing with σ from 0 to an asymptotic value ψ_a , given by

$$\psi_a \exp(-\psi_a) = \delta \quad (21)$$

if δ is smaller than a critical value, $\delta_c = 1/e$. For super-

* This equation could also be obtained from Eq(3), written in terms of the variable ψ , by neglecting the heat conducted $\partial\psi/\partial x$ to the solid and by linearizing the Arrhenius exponent.

supercritical values of δ , $\delta > 1/e$, ψ_0 grows from 0 to 1 when σ grows from 0 to a critical value σ_c given by

$$\delta \exp(-1/\sqrt{\sigma_c}) = 1/e \quad (22)$$

In this supercritical solution when σ approaches σ_c , $\psi_1 \rightarrow \infty$, because $\psi_0 \rightarrow 1$ and $d\psi_0/d\sigma \rightarrow \infty$. The expansion in Eq(18) fails and should be replaced by another involving the stretched variable $(\sigma - \sigma_c)\alpha^{3/2}$. The first approximation for ψ in this transition stage shows a thermal runaway at an ignition time different from σ_c by an amount of order $\alpha^{-3/2}$. Thus Eq(22) can be used in first approximation to calculate the ignition time for supercritical values of δ . In terms of the original variables the ignition time t_i is given by the relation

$$\theta_a(1-\theta_i)/\sqrt{\pi t_i} = \ln\{e\theta_a B e^{-\theta_a}\} \quad (23)$$

For subcritical values of δ , that is for values of B smaller than a critical B_c ,

$$B_c e^{-\theta_a} = 1/e\theta_a \quad (24)$$

no ignition time, leading to high temperatures exists,

the chemical reaction proceeds at an asymptotic temperature given by Eq(21) which is close to the source temperature.

V CONCLUSIONS

A closed form relation has been found between the nondimensional frequency factor B and the ignition time t_i by solving, for large values of the nondimensional activation energy, the equations (1)-(3), or the integral equation(4), which describe the heterogeneous ignition under convective heating by a hot gas.

The analysis sketched in Section III led to the closed form relation(9) for the ignition time for values of B such that relation(7) is satisfied. The analysis of Section IV corresponds to values of B such that δ , given by Eq(10), is of order unity; the analysis indicated the existence of a short inert stage where the surface temperature is given by Eq(6), followed by a long reactive stage when the surface temperature is given in first approximation by

$$\theta = 1 + \theta_a^{-1} (\psi_0 - 1/\sqrt{\sigma}) \quad (25)$$

and the nondimensional increment in surface temperature ψ_0 due to the reaction, is given by Eq(19), with

$$\sigma = \pi t / \theta_a^2 (1 - \theta_i)^2 \quad (26)$$

For values of δ below a critical value $\delta_c = 1/e$, ψ_0 grows asymptotically to a value ψ_a given by Eq(21), while for larger values of δ a thermal runaway is found at ignition time t_i given by Eq(23).

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